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To: Dr. D. Mills
Director of Research

TRIP REPORT: PAPER ON LASER TRITIUM SEPARATION PRESENTED AT THE CONFERENCE ON LASERS AND ELECTRO-OPTICS SAN FRANCISCO, JUNE 9-13, 1986

Hugh D. Morrison

A computer model of a waveguide cell for laser tritium separation was presented in a paper at the 1986 joint meeting of the Conference on Lasers and Electro-Optics and the International Quantum Electronics Conference. The sessions on laser chemistry covered isotope separation of tritium, uranium and zirconium, and production of materials such as ceramics. Many other topics related to lasers and fiber-optics were discussed at the meeting, including laser fusion, laser welding and cutting, and many diagnostic techniques. Technical digests summarizing all the proceedings of both conferences are available. The commercial exhibition accompanying the conferences proved very useful for updating information on laser products and for establishing personal contact with company representatives.

INTRODUCTION

This year San Francisco was the site of the largest ever assembly of laser scientists and engineers at the joint meeting of the Conference on Lasers and Electro-Optics (CLEO) and the International Conference on Quantum Electronics (IQEC). Over 6700 people attended the technical sessions of 917 papers and the exhibits displayed by 282 companies. The conferences were opened by a joint session of plenary papers and award presentations sponsored by the Optical Society of America and by the Institute of Electrical and Electronics Engineers. The first plenary paper was an overview of the Hubble Space Telescope, describing its design and potential for stellar observation. Unfortunately, plans to launch this telescope into earth orbit have been delayed indefinitely due to the recent space shuttle disaster. The other three plenary papers reviewed the status of free-electron lasers, the new field of single atom laser spectroscopy, and the promise for advances in laser surgery.

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CLEO was the forum for research and development in all types of lasers and electro-optic devices with an emphasis on engineering and application. In contrast IQEC covered all aspects of quantum electronics with an emphasis on fundamental physics. The sessions in both conferences were interesting but there were no really exciting new developments in any subject field. I found that the exhibits were as interesting as the sessions so I spent most of one day in the exhibit hall, speaking with representatives of companies whose products we purchase and learning about the many other companies and products in the laser industry. My overall impression supported by other attendees, was that the conferences this year were worthwhile for all concerned but that they have become too big and too diverse to allow adequate time between the many sessions and exhibits for attendees to meet and discuss among themselves.

LASER CHEMISTRY

One of the sessions on Laser Chemistry was titled "Materials Production and Modification". The first presentation was an invited paper delivered by Dr. George Paul of the University of New South Wales, Australia. His talk began with a colour slide show of Australia to entice people to visit and proceeded from there to discuss the operation of the new institute for laser applications of which he is the director. He listed four projects that are ongoing and elaborated on two of them: synthesis of ceramic powders and ultra-violet curing of polymers. The project on powder synthesis is being managed by Dr. Jim O'Neill who is on attachment there from Ontario Hydro Research Division for one year. These projects were aimed at examining areas where conventional sources of ultraviolet light, mercury lamps, could be replaced by excimer lasers. Although the institute was initially funded mainly by the government, it is expected that the active and growing participation by industry will gradually supplant most of the government support.

Three of the remaining four papers in this session discussed isotope separation. The first of these was on tellurium which has importance as a precursor for producing iodine-123 used in nuclear medicine. The last two papers examined tritium separation: one was the paper that I presented and co-authored with Dr. Jim O'Neill, and the other was by Dr. Kazuo Takeuchi of the Institute of Physical and Chemical Research in Japan. The paper by Dr. Takeuchi presented detailed results obtained with a multi-pass photoreactor for multiphoton dissociation of chlorotetrafluoroethane. He reported a selectivity of 2000 for tritium from hydrogen with 80% depletion in continuous flow with a laser input of 2 J at 5 Hz. These results are impressive but his choice of molecule will probably complicate the chemical exchange step and the separation step in a complete separation process as compared to using trifluoromethane (CF_3H) as the process molecule. Tritium selective dissociation of CF_3H was first demonstrated by Dr. Takeuchi in 1979 and it is the molecule chosen by Ontario Hydro Research Division to demonstrate tritium separation.

I learned later from Dr. Takeuchi that his research group will now be spending most of its time on uranium separation. A discussion with Dr. Irving Herman of Lawrence Livermore National Laboratory (and now with Columbia University) revealed that all work on tritium separation at Livermore had ended two years ago and that since then his work in this field has been limited to writing review papers. Thus, it would appear that Ontario Hydro remains as the only major organization still involved with the development of a laser process for tritium separation.

PAPER PRESENTATION

The paper that I presented describes a computer model of a waveguide cell for laser isotope separation of tritium based on multiphoton dissociation of CF_3T by a CO_2 laser. (A copy of the text and slides for the paper is attached as Appendix A.) The model assumes that the pulse of

laser energy is a gaussian shape in space and that the CF_3T is only dissociated for energy fluences that exceed a certain threshold. Input to the model includes values for parameters describing the physical conditions of the gas in the cell, the cell geometry and material properties, the laser wavelength, and the dissociation threshold. A "benchmark" set of parameters was chosen based on a previous order of magnitude design for a laser detritiation plant processing 100 litres per day of tritiated light water to remove 99% of the tritium. Some of the parameters are based on available experimental data (eg dissociation threshold), while other parameters reflect design decisions for a practical plant (eg the number of lasers and waveguides). Although the benchmark values almost completely describe the dissociation cell, there are two independent parameters that can be varied to yield the desired detritiation factor with a minimal consumption of laser energy. These independent parameters, namely the radius of the waveguide and the radius of the laser pulse, control the coupling of the laser energy into the waveguide and the manner in which that energy travels through the waveguide. (The length of the waveguide is also variable but it is set by the point where the energy fluence, reduced by attenuation, falls below the dissociation threshold.) The optimal design for the benchmark case is a cell 20 m long and 2.5 mm in radius and requires an average laser power of 312 W operating at 50 Hz. The full-scale plant would require 80 dissociation cells and lasers of this size to process 100 litres per day of tritiated water.

The individual effect of several input parameters on the optimal design was studied by varying only one parameter from its benchmark value and then optimizing the two independent parameters. One surprising result was that in every case the ratio of the beam radius to the waveguide radius converged to a single optimal value, corresponding to a fixed pattern of coupling energy into the waveguide. This will greatly simplify the design of optics for any size of dissociation cell. The remaining results of the parametric study illustrate that a wide range of design choices is available to meet the engineering requirements of a full-scale detritiation plant with the present technology of lasers, optics and waveguides. This simple computer model has been very useful in assessing the problem of laser tritium separation in waveguides and will continue to be applied in designing further experiments to improve our knowledge of this laser process. The paper was well received and prompted several technical questions and a few requests for copies of the paper.

URANIUM AND ZIRCONIUM ISOTOPE SEPARATION

One session of four papers was devoted to isotope separation of uranium and zirconium. The first paper was an invited presentation on the status of Atomic Vapour Laser Isotope Separation (AVLIS) for uranium at Lawrence Livermore National Laboratory. The US Department of Energy announced last year just after CLEO'85 that they had selected AVLIS over all other methods as the process for separating uranium. The paper this year was an overview that included few technical details and reported that the projected cost for uranium production by AVLIS had remained relatively constant during the last five years at $25/SWU (Separative Work Unit is a measure of uranium separation).

The second paper was an invited talk from Uranit, a West German company, presenting their laboratory scale work on uranium separation based on multiphoton excitation of UF_6 with continuously tunable 16-micron laser systems. They see this work as the development of a process that may be implemented within the next ten years as an adjunct to their present production method of separating uranium by diffusion.

Another paper in this session presented work performed at Isotope Technologies in the US that claimed a photochemical process for uranium separation costing only $10/SWU. This process was based on a chemical reaction of UF_6 with HCl induced by a CO laser. It will be interesting to see if this process can compete with AVLIS now that the Department of Energy has made its choice.

The lone paper on zirconium separation originated from Westinghouse and reviewed the fundamentals of the photo-assisted chemical exchange process developed there. There was no news here as this information had been presented to Ontario Hydro a year before at our own Isotope Separation Workshop. The zirconium separation programs at Westinghouse are still on hold and are expected to remain that way until a major demand for denatured zirconium arises. Since the major expected use of denatured zirconium is for pressure tubes in CANDU and other heavy-water reactors, it is unlikely that demand will increase until some denatured zirconium is available for testing.

OTHER SESSIONS

Time limitations and overlapping of sessions did not permit me to attend the sessions discussed below, but since they are of general interest they are included here. I have copies of the technical digests of both conferences which I will loan to anyone interested in more details on any of the topics presented.

Laser fusion continues to be a very active area and five sessions were devoted to it and related topics such as laser-produced plasmas. The many papers included reports on milestone developments in short-wavelength laser fusion performed with the NOVA laser at Lawrence Livermore National Laboratory, GEKKO XII at Osaka University, and the OMEGA laser at the University of Rochester Laboratory for Laser Energetics. Over the last year there has been a nearly 400 times increase in thermonuclear energy released in laser fusion experiments. Many advances in high power Nd:glass, KrF and iodine lasers for fusion experiments were also reported.

There were six sessions on Industrial Applications of lasers. The topics included the use of lasers for welding and cutting, laser measurement techniques in high temperature gases, and flow diagnostics. One session consisted entirely of papers on fiber-optic sensors that could, for example, sense temperature or the presence of chemicals.

OTHER CONTACTS

There was a poster paper from McMaster University by a student of Dr. John Reid on detecting HTO with a tunable diode laser system. They have achieved a detection sensitivity of 4 ppb in air by a technique that freezes out some water from the air and then releases the water as vapour into a low-pressure chamber. The manufacturer of the tunable diode lasers is the Laser Analytics Division of Spectra-Physics. The basic price for such a laser is US$40,000, and there are many add-ons available for additional cost to provide a complete laser monitoring system.

In the paper that I presented the benchmark value for the damage threshold of the cell windows is taken as 10 J/cm^2, and is allowed to vary down to 0.2 J/cm^2. The study shows that thresholds as low as 1 J/cm^2 do not require any significant increase in laser power. In talking to representatives of several window manufacturers I learned that the highest damage threshold for commercially available windows has remained constant at 4.5 J/cm^2 over the last few years for windows from Harshaw, and windows having damage thresholds of 1 J/cm^2 are readily available at lower cost. Thus, no major developments in this field are required to ensure the success of our tritium separation process.

I met with Mr. Thomas Chisholm of Infrared Industries and we discussed the design for a two-colour detector to be used in a line temperature sensor project at Research Division. Infrared In-

dustries manufactures two-colour detectors containing Si with PbS and Si with PbSe. I have tested both of these detector types and have decided that a two-colour detector containing PbS with PbSe is required. Since this is an unusual combination of detectors and a custom item, the cost will be about $500, as compared to $110 for their regular detectors.

I also met with Mr. Philip Marlowe of Cleveland Crystals to check on their progress in manufacturing crystals of $AgGaSe_2$, which is used for frequency doubling the output of CO_2 lasers. Our interest is in testing for multiphoton dissociation of CF_3T near wavelengths of 5 microns. According to Mr. Marlowe they are having difficulties in manufacturing $AgGaSe_2$ and furthermore they must fulfill government contract commitments for this material before they can begin to fill commercial orders.

SUMMARY

The joint CLEO and IQEC conference of 1986 was the largest ever meeting of laser scientists and engineers. The technical programs and commercial exhibits were all very interesting but the many parallel sessions made it impossible to attend as many papers as I would have liked. The application of lasers to photochemistry and isotope separation as well as industrial uses such as welding, cutting and remote diagnostics continue to be important areas of research and development in the laser community. However, the major interest in laser isotope separation has shifted from fundamental research to engineering application over the last few years and has become concentrated into a few large research centres. The paper I presented describing a computer model of a waveguide cell for tritium separation was only one of two papers about tritium separation and was well received, prompting several requests for copies. Several new contacts were made and old ones were renewed, thus making this conference all the more worthwhile.

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APPENDIX A

OPTIMAL CELL DESIGN FOR LASER TRITIUM SEPARATION

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Slide 1: At Ontario Hydro we have developed a complete process for laser isotope separation of tritium based on multiphoton dissociation of CF_3T (trifluoromethane) with high power CO_2 lasers [1]. CF_3T has two big advantages: The first is that on dissociation there is only one tritium containing product, ie TF. The second advantage is that TF can be removed from the process stream by using a sodium fluoride filter, with no energy cost. Because of these advantages and other favourable chemical properties, we have been able to develop a complete process cycle for decontaminating tritiated light water using trifluoromethane as the process gas [2]. The one disadvantage of CF_3T is that the energy fluence for dissociation exceeds the damage threshold of windows and mirror materials. The simple solution is to focus the laser energy into a large cell. But we have demonstrated that a waveguide cell is much more efficient [3]. In this paper I will describe a model of our waveguide cell and illustrate how its design can be optimized for minimum laser power. This model will eventually be integrated into a model for the whole process that will be used to optimize the design of full-scale separation plants.

Slide 2: The waveguide itself is a hollow cylindrical glass tube of radius "a", with larger diameter end pieces closed by windows. The input laser beam is taken as a linearly polarized gaussian, focussed through one window to a beam waist of radius "w" at the waveguide entrance so that the peak intensity is the threshold for dielectric breakdown. The ratio "w/a" determines how the laser power is coupled into the waveguide modes. A linearly polarized gaussian beam only couples into the linearly polarized, radially symmetric EH_{1m} (m=1,2,...) modes [4].

Slide 3: If we look at the power coupled into the EH_{1m} modes as a function of "w/a" we see that the EH_{11} mode receives the greatest power over the largest range. Note also that the peak in the EH_{11} mode power coincides with the beginning of the decline of total power coupled into the waveguide. The region of the dotted box is shown in greater detail in the next figure to illustrate the power in the higher modes around this peak in the EH_{11} mode.

Slide 4: Here we see that the EH_{12} mode contains no power for "w/a" about 0.7, and that only ~1% of the total power lies in higher order modes. Preliminary calculations with our model treated power coupling into the lowest two modes only. It quickly became apparent that the optimum coupling for maximum dissociation occurred when the EH_{12} coupling coefficient was zero.

Slide 5: To understand why zero EH_{12} coupling is the optimum coupling we must look at the intensity distribution in the waveguide and at the theoretical model assumed for dissociation. In our model, the dissociation probability as a function of energy fluence is a step-function. That is to say that all molecules that see a fluence greater than a threshold value are dissociated and the rest are unaffected. In this diagram the dashed horizontal line represents the dissociation threshold, and the peak intensity is set to the threshold for dielectric breakdown. For a pure EH_{11} mode, all molecules within the radius of intersection of the solid curve and the dashed horizontal line are dissociated. If power is also coupled in the EH_{12} mode then the resulting in-

tensity distribution must be obtained by summing the electric fields of EH_{11} and EH_{12}, as given in the top panels, and normalizing the peak to the threshold for dielectric breakdown. The maximum intensity occurs when the two modes are in phase (upper dashed curve), which produces a smaller radius for dissociation and fewer molecules dissociated. Since the modes have different phase velocities they will move out of phase (lower dashed curve) and the peak intensity can drop below the dissociation threshold farther down the waveguide. Thus the maximum volume for dissociation occurs when only the EH_{11} mode is excited in the waveguide.

Slide 6: To demonstrate the use of the computer model we selected a set of input parameters appropriate to a plant that removes 99% of the tritium from a water stream of 100 l/day. These values represent our "benchmark" case for a typical application of tritium separation. The input parameters fall into four categories. The gas parameters describe the conditions of the trifluoromethane gas in the dissociation cell and list the best known values for the breakdown and dissociation thresholds. One of the waveguide cell parameters is the window damage threshold. It determines how close the window can be to the waveguide. The length and radius of the waveguide are not listed here because they are variable parameters optimized by the program. The laser is a CO_2 laser tuned to 9.3 microns, running at 50 Hz. The pulse energy is fixed by the waveguide radius, by the beam coupling parameter and by the condition that the peak intensity at the waveguide entrance is the dielectric breakdown threshold. Among the system parameters, the gas flow is the total trifluoromethane equivalent of 100 l/day of water. We assume that there are 80 modular units in the plant, that is 80 lasers and 80 waveguides operating in parallel, so that each waveguide processes 1/80 of the total gas flow. In each waveguide 99% of the tritium is removed because the gas sees many laser pulses as it flows through a waveguide.

Slide 7: For the input values of the previous table, the computer model produced the output values shown here, optimized for minimum laser power at the required depletion factor of 99%. The optimal waveguide dimensions are practical and realistic: 0.25 cm in radius, 14.4 m long with window sections 3.6 m long for a total cell length of ~21 m. This size of cell requires a laser pulse energy of 6.2 J, which means an average laser power of 312 W at 50 Hz. Such a laser is certainly possible with present technology. Note that this is the minimum laser power required by each of the 80 parallel modules to achieve a depletion of 99%. I will now briefly show how that minimum power is changed as several input parameters in the benchmark case are varied one at a time. Keep in mind that for a fixed coupling parameter the laser power scales with the square of the radius of the waveguide.

Slide 8: First - what laser power do we need to increase the depletion? A factor of 100 greater depletion from 10^{-2} to 10^{-4} remaining tritium fraction requires only twice as much laser power. This corresponds to increasing the waveguide radius by 40% for the same cell length. That is a relatively inexpensive way to increase depletion provided laser costs scale linearly with power. We will now fix the depletion at 99% and vary some of the other parameters.

Slide 9: This slide and the next examine the effect of the two important fluences in the model: the breakdown threshold and the dissociation threshold. These are not variable parameters as such, but their values are not well known. If experiments show that these two parameters lie closer together than the selected values of 100 and 65 J/cm^2, then significantly higher laser powers will be required. If they lie farther apart then much less power will be needed to achieve a given depletion. This graph shows the effect of fixing the dissociation threshold and varying the breakdown threshold above 65 J/cm^2.

Slide 10: In this second graph the breakdown threshold is fixed at 100 J/cm^2 and the dissociation threshold is allowed to vary. Again note that as these two fluences approach each other in value, much higher laser powers are required.

Slide 11: Now let us examine the length of the waveguide. The optimum waveguide length for the benchmark case is ~14 m. Beyond that length no dissociation occurs because at that point the peak fluence in the waveguide is less than the threshold for dissociation. Thus longer waveguides use no more laser power to achieve a depletion of 99%. On the other hand, shorter lengths can be employed to the same effect by using more power in a larger radius waveguide. For lengths down to 5 m, very little increase in power is required.

Slide 12: The overall length of the waveguide cell includes the window sections. The gas in the entrance window section absorbs laser power, and so the power at the waveguide entrance is less than the power at the window. Lower window damage thresholds require longer window sections, and thus more laser power is required to compensate for the absorption loss. The power required for a window loading of 1 J/cm^2 is still tolerable, and such windows are readily available. Higher window loadings, such as the 10 J/cm^2 chosen for the benchmark case are a tradeoff between cheaper lasers and more costly windows. An alternative is to use a more complex cell design so that a non-absorbing gas, such as helium, fills the window section, because then the laser power is independent of window damage threshold.

Slide 13: The final parameter we examine here is the repetition rate of the laser. Surprisingly, the average required laser power is practically independent of the repetition rate. Higher repetition rate means less energy per pulse and smaller radius waveguides. Lower repetition rate implies greater energy per pulse and larger radius waveguides. The dramatic increase in power at the very low repetition rates is due to a rapid increase in window section length.

Slide 14: In summary, we have explored a model of a waveguide cell for trifluoromethane dissociation at high fluences as is needed for tritium separation. The optimum coupling is such that only the lowest order waveguide mode is excited. Using the best available information and a simple threshold model for dissociation, the minimum laser power demands cells 5-15 m long and 2-4 mm in radius; dimensions that are realistic and practical. The laser requirements for a full-scale plant can be met by present technology and flexibility in design choices. More detailed designs will require further dissociation experiments in waveguides and corresponding refinements of the model.

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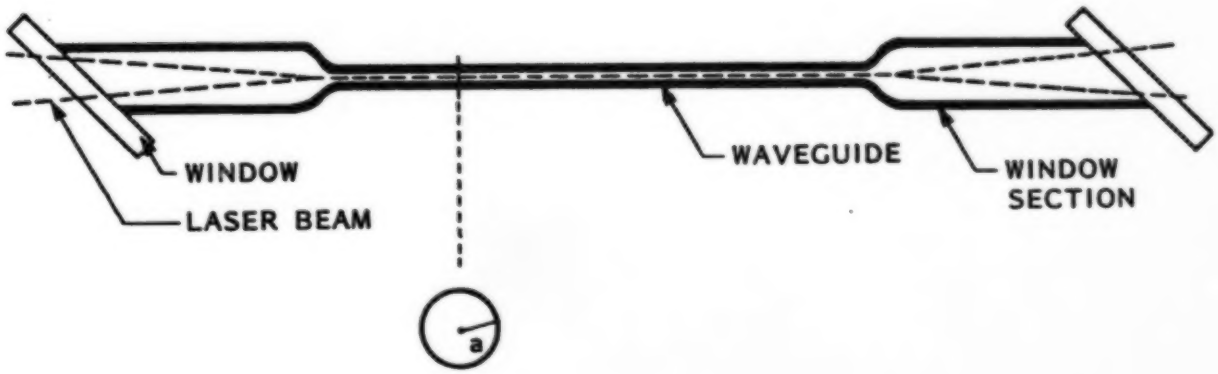
4. E.A.J. Marcatili and R.A. Schmeltzer, "Hollow metallic and dielectric waveguides for long distance optical transmission and lasers," Bell Syst. Tech. J. 43, 1783-1809 (1964).

SLIDE 1:

LASER TRITIUM SEPARATION AT ONTARIO HYDRO

- Tritium separation by multi-photon dissociation of CF_3T
- Complete separation process developed for CF_3H/CF_3T
- CF_3T dissociation fluence exceeds mirror damage threshold
- Waveguide cell performs better than simple focussing
- Optimal cell design required for costing full-scale plants

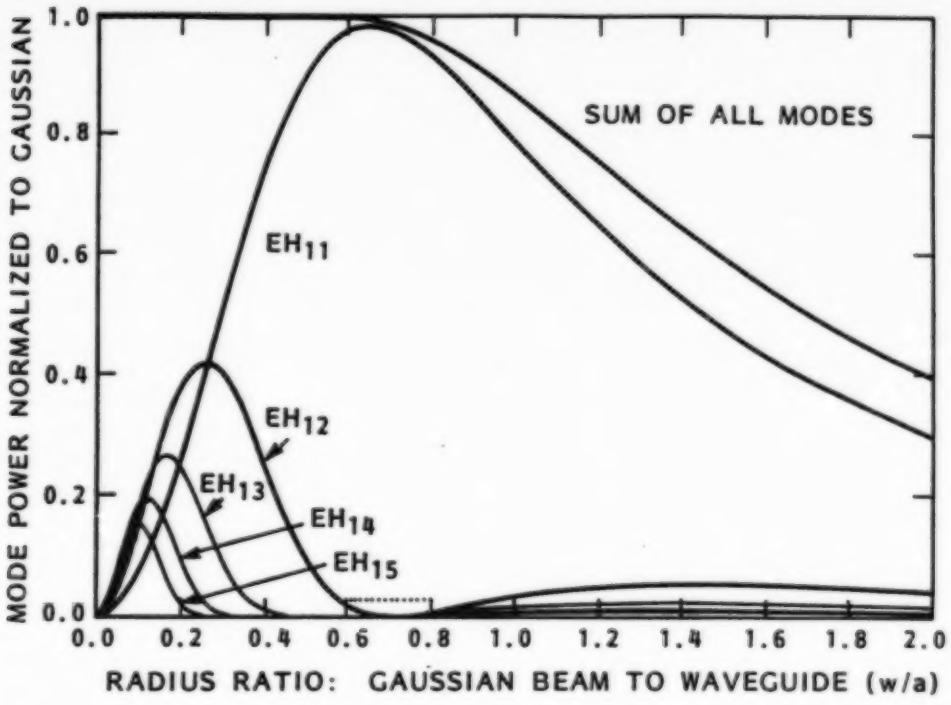
SLIDE 2: **WAVEGUIDE DISSOCIATION CELL**



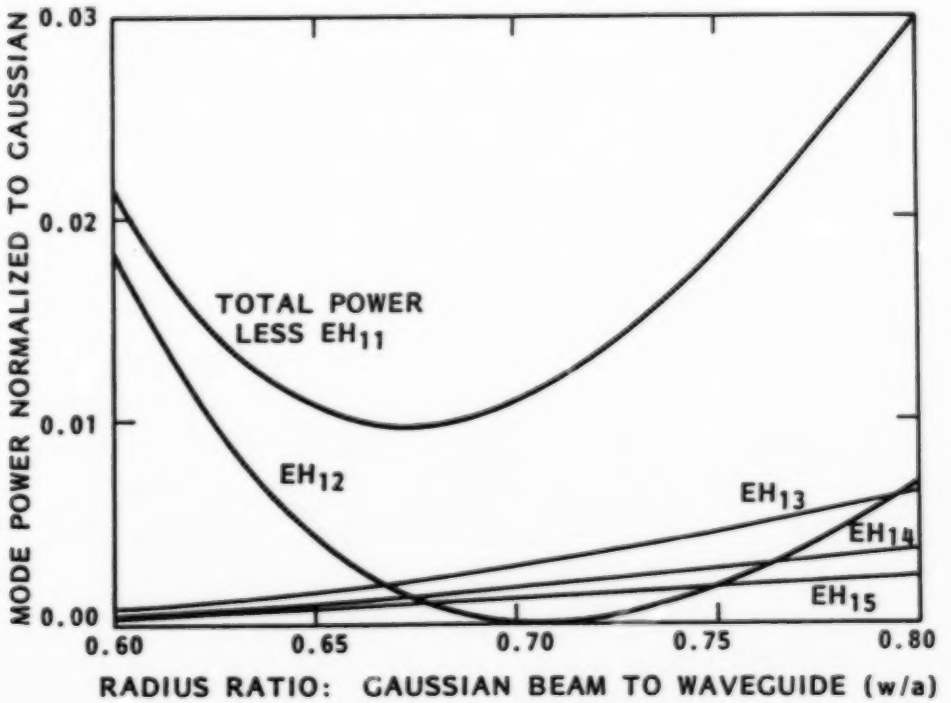
LASER BEAM INTENSITY: $I_o \exp(-2r^2/w^2)$

BEAM COUPLING PARAMETER: w/a

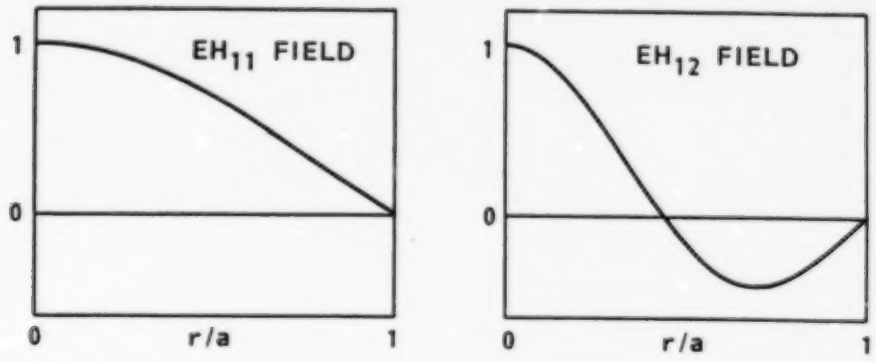
SLIDE 3: **POWER COUPLED INTO WAVEGUIDE**

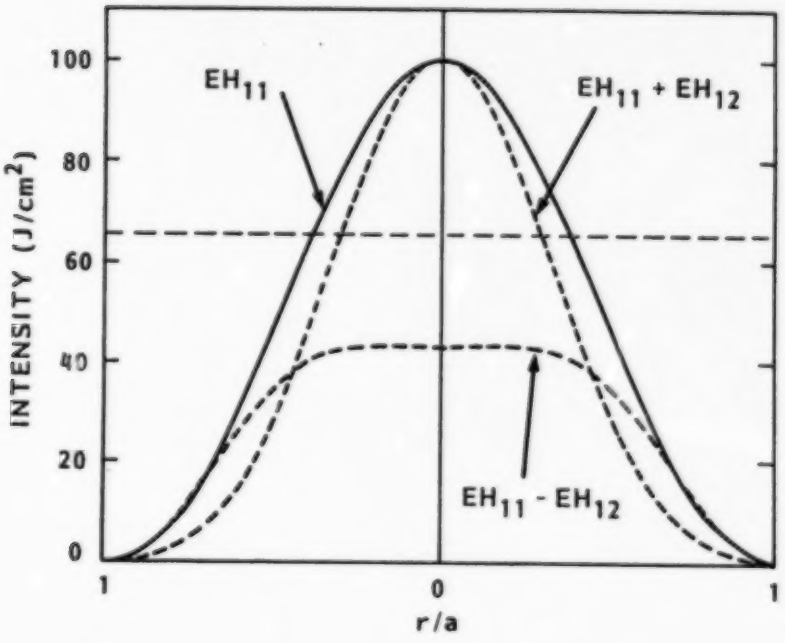


SLIDE 4: **POWER COUPLED INTO WAVEGUIDE**



SLIDE 5: WAVEGUIDE MODE SUPERPOSITION





SLIDE 6: BENCHMARK LASER DISSOCIATION CELL PARAMETERS

INPUT VALUES

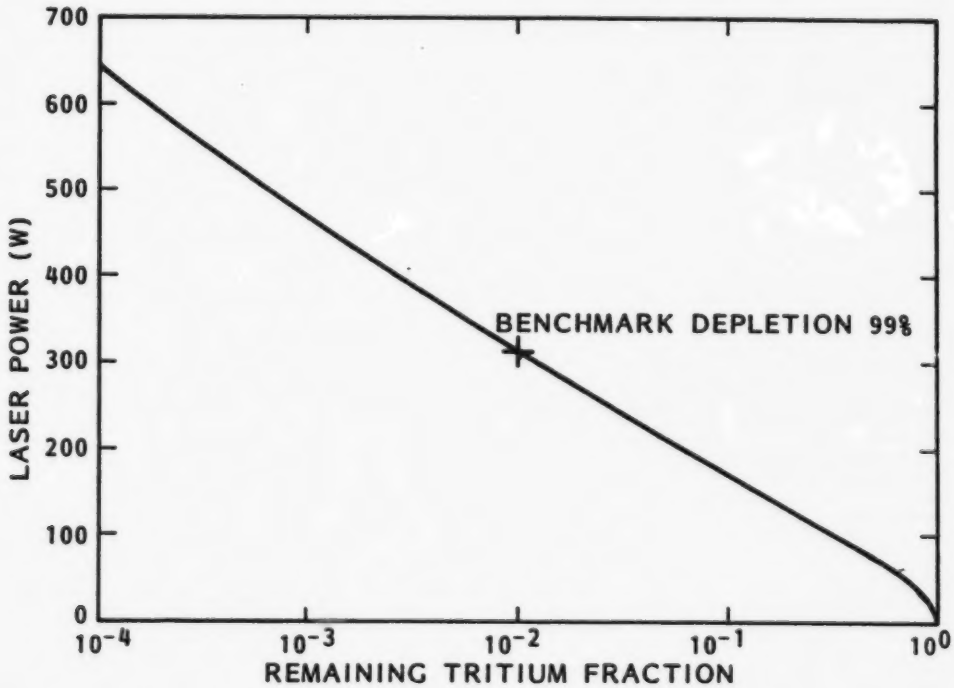
GAS:	Absorption Coefficient	3.17×10^{-6} $cm^{-1}.torr^{-1}$
	Pressure	85 torr
	Temperature	194 K
	Dielectric Breakdown Threshold	100 $J.cm^{-2}$
	CF_3T Dissociation Threshold	65 $J.cm^{-2}$
WAVEGUIDE:	Window Transmission	0.95
	Window Damage Threshold	10 $J.cm^{-2}$
LASER:	Wavelength	9.3 μm
	Repetition Rate	50 Hz
SYSTEM:	Required Depletion Factor	0.99
	Gas Flow	0.129 $mol.s^{-1}$
	Number of Lasers and Waveguides	80

SLIDE 7: BENCHMARK LASER DISSOCIATION CELL PARAMETERS

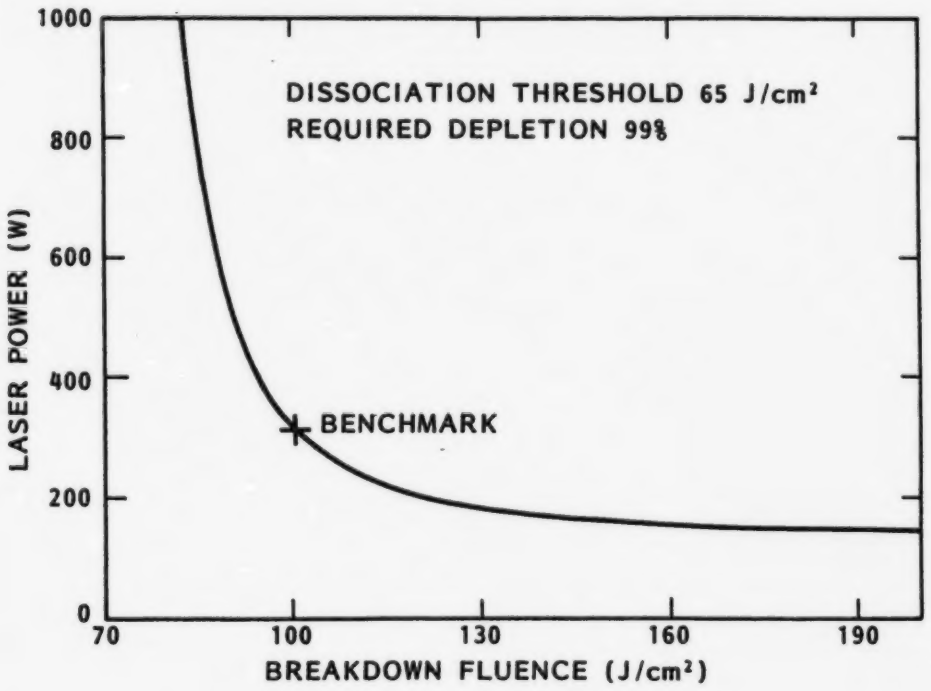
OPTIMAL OUTPUT VALUES

Radius Ratio (Beam/Waveguide)	0.7063
Waveguide Radius	0.249 cm
Waveguide Length	14.4 m
Minimum Waveguide-Window Separation	3.6 m
Laser Beam Radius at Window	0.619 cm
Laser Pulse Energy	6.24 J
Laser Power	312 W

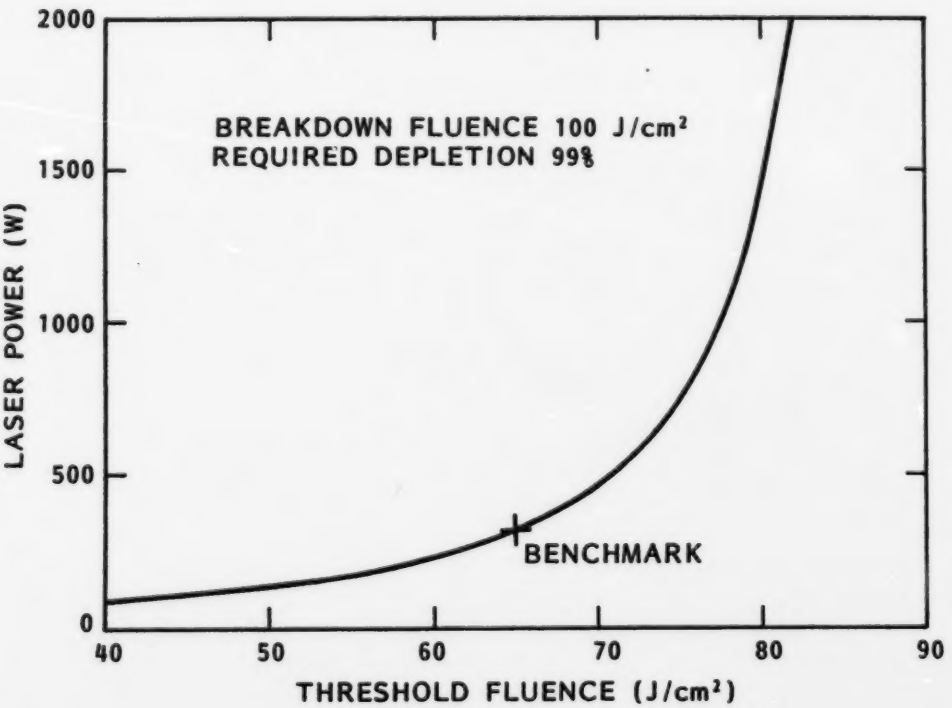
SLIDE 8: **EFFECT OF REQUIRED DEPLETION**



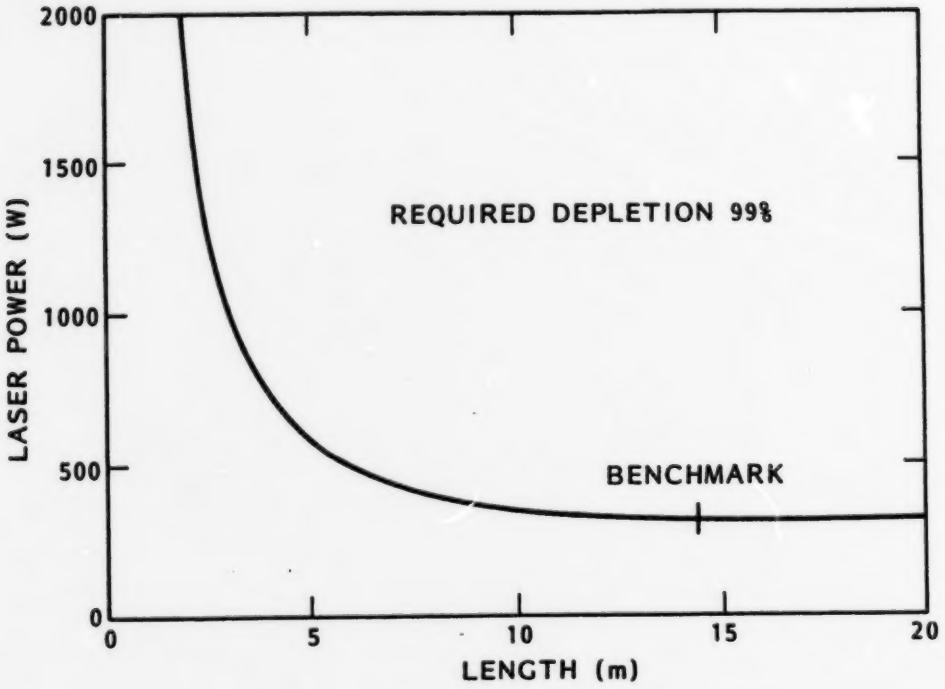
SLIDE 9: **EFFECT OF GAS BREAKDOWN**



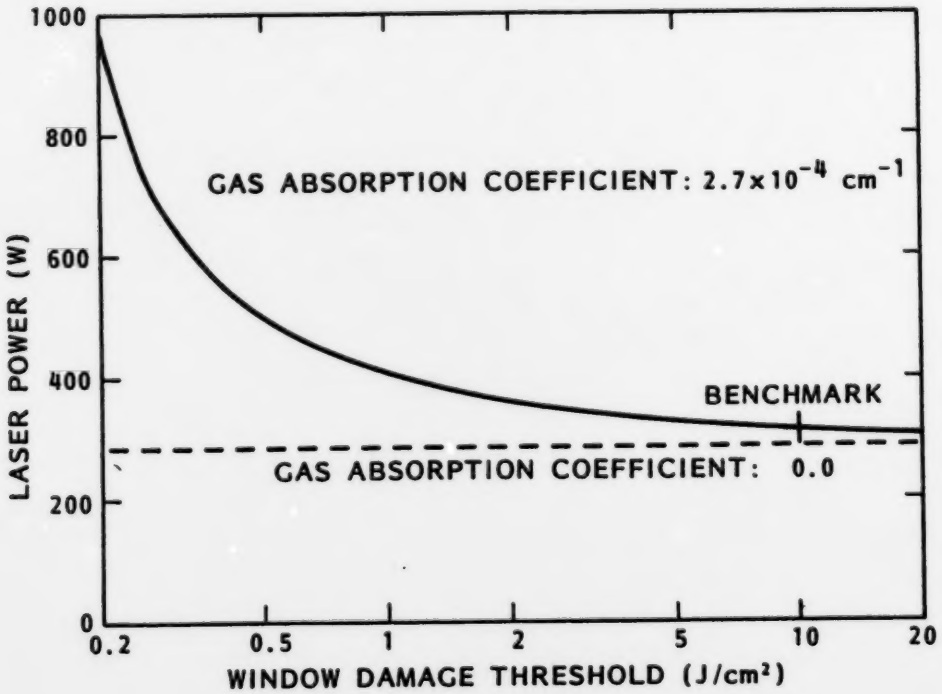
SLIDE 10: **EFFECT OF DISSOCIATION THRESHOLD**



SLIDE 11: EFFECT OF LENGTH OF WAVEGUIDE



SLIDE 12: EFFECT OF GAS IN WINDOW SECTION



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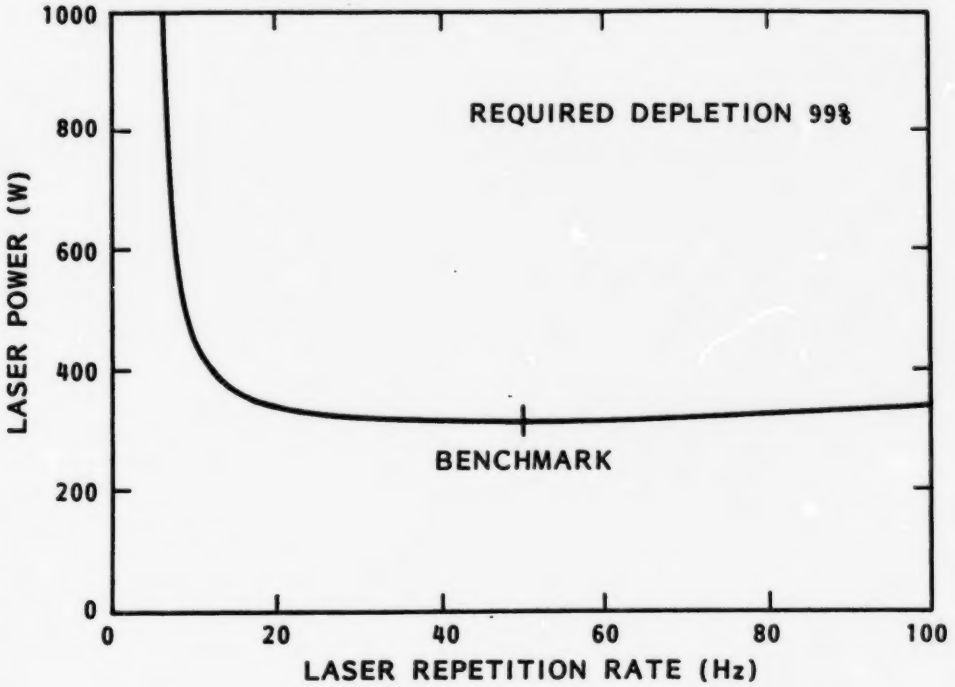
SLIDE 13: **EFFECT OF PULSE REPETITION RATE**



SLIDE 14:

SUMMARY

- **Optimum coupling: w/a = 0.706**
- **Optimal waveguide cells: 5-15 m long, 2-4 mm radius**
- **Laser requirements: technology readily available**
- **Dissociation process: experiments needed to refine model**